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AIR POLLUTANTS: REGIONAL TRANSPORT,
PROPERTIES AND EFFECT ON UPPER TROPOSPHERIC
HUMIDITY

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Academic dissertation

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Laura Irmeli Riuttanen
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Abstract

Climate change and air quality are the big challenges of our time. In this work, air pollution transport to Finland, air pollution properties, their direct radiative effect on climate as well as their effect on upper tropospheric humidity are studied by using satellite data of aerosol optical properties, precipitation, clouds and upper tropospheric humidity, in situ measurement data of gases and aerosols at three measurement stations, combined with flight measurements, air mass trajectories, synoptic maps and meteorological reanalysis data.

South-eastern transport routes with continental origin were found on average to bring the highest concentrations of anthropogenic gaseous and particulate pollution to southern Finland. The air masses coming from the north-east brought air that was clean of anthropogenic pollution but included high concentrations of nucleation and Aitken mode particles. Air mass trajectory statistical methods were found to be useful tools to study air pollution transport, but they were not able to reproduce emission source maps.

The most severe air pollution cases in the history of air pollution measurements in Finland were produced by the biomass burning events in Eastern Europe in 2006 and 2010. The smoke was transported hundreds and even thousands of kilometres and the smoke particles were observed to grow in size during transportation.

Particulate air pollution in southern Finland scattered solar radiation and had a cooling effect on climate. High absorption coefficients were measured in continental air pollution from Eastern Europe as well as in biomass burning smoke, but also these air pollutants were on average more scattering than absorbing.

Based on satellite observations we were able to show the first observational evidence, with a strong emphasis on causality, that aerosols increase upper tropospheric humidity and have thus a so-far overlooked warming effect on climate. The magnitude of the observed increase is so big that the mechanism potentially has relevance for the global climate.

Keywords: air pollution transport, aerosol–cloud interactions, upper tropospheric humidity

Contents

1	Introduction	5
2	Theory	8
2.1	Air pollutants and their sources	8
2.2	Air pollutants and climate	10
2.2.1	Basic concepts of aerosol optical properties	14
2.2.2	Aerosols affect upper tropospheric humidity	15
2.2.3	About meteorological covariation in aerosol–cloud interaction studies	18
3	Research methods	21
3.1	Ground-based measurements of gases and aerosols	21
3.2	Satellite-based measurements	22
3.3	Trajectory analysis	25
4	Results	28
4.1	Transport and properties of air pollutants in Finland	28
4.1.1	Optical properties	30
4.1.2	Biomass burning episodes	31
4.2	Direct effects of air pollutants on climate by scattering and absorption of solar radiation	32
4.3	Indirect effects of air pollutants via changes in cloud microphysics: aerosols increase upper tropospheric humidity	33
5	Review of papers and the author’s contribution	35
6	Conclusions	36
	References	37

Nomenclature

Symbol	Explanation
α	Ångström exponent
σ_{SP}	scattering coefficient
σ_{AP}	absorption coefficient
σ_{EP}	extinction coefficient
AOD	Aerosol optical depth
AR5	IPCC Assessment Report 5
BC	Black carbon
CCN	Cloud condensation nuclei
CS	Condensation sink
ERF	Effective radiative forcing
IPCC	Intergovernmental Panel on Climate Change
RF	Radiative forcing
UTH	Upper tropospheric humidity
VOC	Volatile organic compound

List of publications

This thesis consists of an introductory review, followed by four research articles. In the introductory part, these papers are cited according to their roman numerals.

- I** Riuttanen, L., Hulkkonen, M., Dal Maso, M., Junninen, H., and Kulmala, M. (2013). Trajectory analysis of atmospheric transport of fine particles, SO₂, NO_x and O₃ to the SMEAR II station in Finland in 1996-2008, *Atmos. Chem. Phys.*, 13:2153–2164.
- II** Virkkula, A., Backman, J., Aalto, P. P., Hulkkonen, M., Riuttanen, L., Nieminen, T., Dal Maso, M., Sogacheva, L., de Leeuw G., and Kulmala, M. (2011). Seasonal cycle, size dependencies, and source analyses of aerosol optical properties at the SMEAR II measurement station in Hyytiälä, Finland, *Atmos. Chem. Phys.*, 11:4445–4468.
- III** Leino, K., Riuttanen, L., Nieminen, T., Dal Maso, M., Väänänen, R., Pohja, T., Keronen, P., Järvi, L., Aalto, P. P., Virkkula, A., Kerminen, V.-M., Petäjä, T., and Kulmala, M. (2014). Biomass-burning smoke episodes in Finland from eastern European wildfires, *Boreal Env. Res.*, 19 (suppl. B): 275–292.
- IV** Riuttanen, L., Bister, M., Kerminen, V.-M., John, V. O., Sundström, A.-M., Dal Maso, M., Räisänen, J., Sinclair, V., Makkonen, R., Xausa, F., de Leeuw, G., and Kulmala, M. (2016). Observational evidence for aerosols increasing upper tropospheric humidity, *Atmos. Chem. Phys.*, 16:14331–14342.

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1 Introduction

The climate system has faced manifold changes due to the influence of man. Various compounds emitted to the air have changed the radiative transfer, the hydrological cycle, as well as the chemistry of the air. These changes have had severe impacts on ecosystems.

Air pollutants are gaseous or particulate matter emitted to the atmosphere. Some of them, like carbon dioxide (CO_2), act as greenhouse gases and warm the climate system. On the other hand, particulate matter in the atmosphere scatters sunlight, reducing the amount of radiation reaching the surface and thus cooling the climate system. Air pollutants also have manifold indirect effects on the climate system, for example via their effects on clouds. Many air pollutants are harmful to human health (Hoek et al., 2013).

It seems that emission restriction policies and new technologies have been more effective at reducing particulate pollution than the greenhouse gases. The largest impact of aerosols on climate most likely occurred between 1950 and 1970 (Smith and Bond, 2014). The greenhouse gas emissions, however, have not yet clearly decreased (Jackson et al., 2016). Long-term measurements show dramatic changes in atmospheric concentrations; for example, CO_2 concentrations have increased by more than 40 % since 1750, from 280 ppm to over 400 ppm (Betts et al., 2016). This is mainly due to the burning of fossil fuels and changes in land use (IPCC, 2013).

One example of a major source of air pollution is wildfires, which emit large amounts of various compounds into the atmosphere (Andreae and Merlet, 2001). The majority of biomass burning happens in the tropics, but there are severe fire episodes also in boreal zones every summer (van der Werf et al., 2006). The smoke is transported to vulnerable Arctic areas where it amplifies the already severe effects of climate change (Stohl et al., 2007). Forest fire areas as well as emissions are also projected to increase in a warming climate (de Groot et al., 2013).

Air pollutants are a challenging topic to study because of their various sources and composition. Once emitted to the atmosphere, they can travel long distances and transform in physical and chemical processes during transportation. Their interactions with clouds and other meteorological properties are complex (Fan et al., 2016; Stevens and Feingold, 2009).

Observations are key to understanding the atmosphere and climate. As the phenomena are complex, comprehensive measurements and long time series are needed to monitor changes. However, in situ measurements capture atmospheric properties only at one place at a time. With the help of satellite products and atmospheric modelling, more light can be shed on the processes behind the observations. This requires expertise on multiple methods at once.

In this thesis, I study both gaseous and particulate air pollution concentrations in Finland. I study the general features of air pollution transport to Finland, and the severe smoke episodes in 2006 and 2010 as special cases. The optical properties of these air pollutants are studied and their effect on radiative transfer and climate is discussed. A special emphasis is put on the aerosol effect on upper tropospheric humidity (UTH) via changes in the microphysics of deep convective clouds. We show the first observational evidence of the newly suggested mechanism that has a potentially large impact on global climate. Topics covered in this thesis are presented in Figure 1.

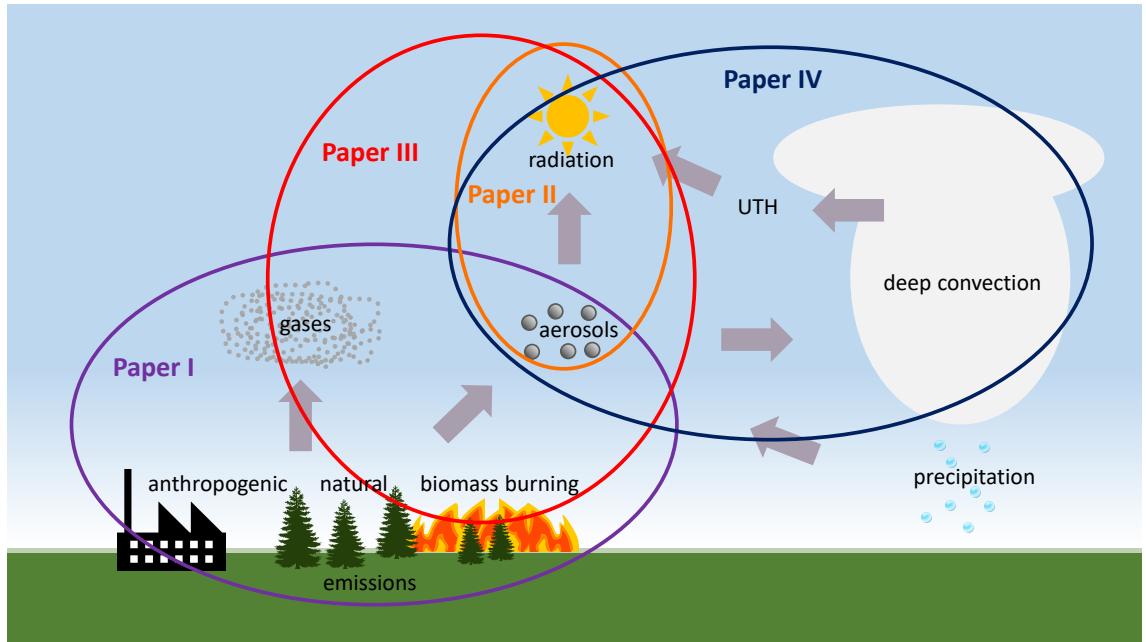


Figure 1: In this thesis I study interactions between gaseous and particulate air pollutants with clouds and climate. Processes covered in this thesis are indicated as arrows. Topics covered in Papers I–IV are presented as circles.

The aims of this thesis are, based on ground-based measurements, remote sensing data and air mass trajectories, to

1. characterize the air pollution measured in remote background air in southern Finland (**Papers I & II**);
2. determine the general characteristics of the origins of these measured air pollutants as well as test the applicability of air mass trajectory statistical methods in reproducing the emission sources (**Paper I**);
3. quantify the effect of biomass burning episodes in 2006 and 2010 on air quality in Finland (**Paper III**);
4. quantify the direct effects of these air pollutants on climate (**Papers II & III**);
5. test the hypothesis of aerosols increasing UTH by using observations (**Paper IV**).

2 Theory

2.1 Air pollutants and their sources

The atmosphere consists of gases and particulate matter. The main constituents are nitrogen (N_2 , 78.1 %) and oxygen (O_2 , 20.9 %; Seinfeld and Pandis, 1998). The remaining 1 % consists of trace gases and particulate matter and their concentrations vary according to sources and sinks. Sources as well as sinks are different for different compounds and they can be both natural and anthropogenic. In addition to that, the atmosphere also contains water in solid and liquid forms in clouds and precipitation: water droplets, ice crystals, snow, hail, graupel – and everything in between.

Carbon monoxide (CO) and **carbon dioxide (CO_2)** are emitted during the burning of any material of organic origin. The more incomplete the burning, the larger is the CO/CO_2 ratio. The main sources of CO are methane oxidation, combustion and industrial processes, and biomass burning (Seinfeld and Pandis, 1998). The main sources of CO_2 are combustion of fossil fuels, cement production and land use changes (Ciais et al., 2013). **Sulphur dioxide (SO_2)** is emitted during combustion of fossil fuels and industrial processes (Smith et al., 2011). **Nitric oxide (NO)** and **nitrogen dioxide (NO_2)** – together referred to as NO_x – are produced in high temperature combustion. The main sources are fossil fuel combustion, release from soils and biomass burning (Seinfeld and Pandis, 1998). **Ozone (O_3)** is formed in the atmosphere in photochemical reactions of NO_x , CO and volatile organic compounds (VOCs, Seinfeld and Pandis, 1998). **Water vapour (H_2O)** is emitted to the atmosphere via evaporation from surfaces. Atmospheric water vapour content is strongly governed by temperature, and therefore it is often given in units of relative humidity (RH).

The liquid and solid particles in the atmosphere are called **aerosol particles** and together with the carrier gas they are called **the aerosol**. Some aerosol particles are emitted directly to the atmosphere, like dust or sea spray lifted by winds, and these are called **primary aerosols**. Also for example pollen and viruses emitted by living organisms are primary aerosols. Particles are also formed from precursor vapors in the atmosphere and these are called **secondary aerosols**. **New particle formation** happens when vapors nucleate in favourable conditions, molecules stick to each other, and surrounding vapors condensate on the surface (Kulmala et al., 2013). Sulphate aerosol forms from SO_2 , nitrates from NO_x , and organic matter from

VOCs – with other compounds included in the formation process. Combustion is a major source of both primary and secondary particles. **Black carbon (BC)** is emitted in combustion processes (Bond et al., 2013). Wildfires are globally a significant contributor to atmospheric aerosol loading.

Aerosol particle sizes range from molecular level (freshly formed clusters in the atmosphere consist of a couple of molecules and can be 1–2 nm in diameter; Kulmala et al., 2013) to tens of micrometres. Dust, pollen or soot particles can be larger than 100 μm , but particles of that size and larger tend to fall to the ground due to gravity and thus do not travel long distances. **Fine aerosol particles** (diameter smaller than 2.5 μm) are usually secondary aerosols, whereas **coarse aerosol particles** (diameter larger than 2.5 μm) are primary aerosols.

Fine aerosol particles are divided according to their diameter into nucleation mode, Aitken mode, and accumulation mode. **Nucleation mode** particles have a diameter smaller than 25 nm. They are freshly formed from precursor gases in the atmosphere. **Aitken mode** particles have a diameter of 25–90 nm. They are particles grown from the nucleation mode. Particles in the **accumulation mode** have a diameter of 90 nm to 2.5 μm . This is the most persistent size mode of atmospheric particles, as particles smaller than that tend to grow and particles larger than that tend to fall from the atmosphere due to gravity. Particles in the accumulation mode can stay in the atmosphere for weeks and travel hundreds or even thousands of kilometres.

The **condensation sink (CS)** describes the effectiveness of surrounding vapors to condensate on existing particles in units of s^{-1} (for formula see Kulmala et al., 2001a). In practice, the more there is pre-existing aerosol to which the vapors can condensate, the faster the vapors stick to the surfaces. The same applies to small particles, too, which tend to coagulate into the pre-existing aerosol. The removal processes are fast for nucleation and Aitken mode particles.

Once emitted to the atmosphere, air pollutants are transported in atmospheric motions, i.e. winds. They are removed from the atmosphere by precipitation (**wet deposition, wet scavenging**) and by falling due to gravity or sticking to the surfaces (**dry deposition**). As air pollutant sources, sinks and transport are strongly affected by meteorology, air pollutant concentrations are always a strong function of meteorological conditions.

As stated above, atmospheric constituents can be of both natural and anthropogenic

origin, and division between them may not be straightforward. For example, forest fires that emit a lot of smoke can be lit by humans or, for example, by lightning. And even if a fire was lit by lightning, there may be anthropogenic factors behind the environmental conditions that favour the spread of the fire, like erosion of land due to land use changes or climate change induced drought. Regarding the fire episodes studied in **Paper III**, the spring 2006 fires were largely lit by agricultural burnings – a traditional habit of land cultivation that is currently forbidden in the European Union but still widely used in Eastern Europe (Stohl et al., 2007, see also Figure 2). Summer 2010 was exceptionally warm in large parts of the northern hemisphere, and as heat waves and summer droughts are also predicted by climate models in these areas in a warming climate, the contribution of global warming cannot be ruled out (Trenberth and Fasullo, 2012).

As **air pollutants** in this thesis I call 1) all aerosol particles in the atmosphere regardless of whether they are of natural or anthropogenic origin, and 2) all gases in the atmosphere except N_2 , O_2 and water vapour (but in this thesis we mainly consider CO , CO_2 , SO_2 , NO_x (NO and NO_2) and O_3).

2.2 Air pollutants and climate

The energetics of our climate system is defined by a balance between incoming solar radiation and outgoing long-wave radiation. The Earth system gets almost all of its energy from the Sun as short-wave electromagnetic radiation. Approximately half of this radiation is absorbed by the surface of the Earth; the rest is absorbed or reflected by the atmosphere and clouds, and a small fraction is reflected by the surface (Wild et al., 2013). The surface of the Earth, as well as the atmosphere and clouds, emit the absorbed energy as long-wavelength thermal radiation.

By altering atmospheric composition or surface properties, humans have changed this radiative equilibrium, pronouncedly during the past two centuries. The magnitude of this change is expressed using the concept of **radiative forcing, RF** ($W\ m^{-2}$). Radiative forcing is defined as *the change in net downward radiative flux at the tropopause after allowing for stratospheric temperatures to readjust to radiative equilibrium, while holding surface and tropospheric temperatures and state variables such as water vapour and cloud cover fixed at the unperturbed values* (Myhre et al., 2013). The Intergovernmental Panel on Climate Change’s (IPCC) Fifth Assessment Report (AR5) also



Figure 2: It was a common tradition also in Finland to burn the land before cultivation until the early 20th century (Soininen, 1959). Source: I. K. Inha (1893), Kaskenviertäjiä Enossa; Museoviraston Kuvakokoelmat.

defines a concept of **effective radiative forcing, ERF**, as *the change in net top of the atmosphere downward radiative flux after allowing for atmospheric temperatures, water vapour and clouds to adjust, but with surface temperature or a portion of surface conditions unchanged* (Myhre et al., 2013).

The response of the climate system to a radiative forcing is described by the concept of **climate sensitivity**. Climate sensitivity is defined as *the change in global mean surface temperature at equilibrium that is caused by a doubling of the atmospheric CO₂ concentration* (IPCC, 2013). According to IPCC AR5, climate sensitivity is likely in the range of 1.5 °C to 4.5 °C (IPCC, 2013).

Some of the anthropogenic pollutants act as **greenhouse gases**. They absorb the

long-wave radiation emitted by the surface of the Earth, thus decreasing the outgoing long-wave radiation. This must be compensated for by increases in surface and lower tropospheric temperatures to again increase the outgoing long-wave radiation. Increased greenhouse gas concentrations, most importantly CO₂, are the main cause of the observed increase in surface temperatures of the Earth since 1750 (IPCC, 2013). Water vapour is also a greenhouse gas. It creates an important feedback mechanism in the climate system as increased surface temperatures tend to increase tropospheric water vapour content (Held and Soden, 2000). However, relative humidity is commonly assumed to stay constant. This assumption is based on climate models that almost uniformly show no changes in average global relative humidity with increasing surface temperature (Held and Soden, 2000), although local changes may exist (Sherwood et al., 2010). In contrast to that we show in **Paper IV** that aerosols may have an anthropogenic effect on **upper tropospheric humidity (UTH)**, i.e. relative humidity of the upper troposphere.

Aerosol particles interact with incoming solar radiation and thus have a direct effect on the radiative balances of the atmosphere and the surface, and thus climate. They can scatter solar radiation into different directions and absorb it. Scattering and absorption depend on aerosol properties, for example particle composition and size. For example, aerosols containing BC absorb sunlight, whereas ammonium sulphate aerosols can be considered as purely scattering (**Paper II**). Scattering aerosols cool the surface, whereas absorbing aerosols warm the atmospheric layer they are located in. Interaction with solar radiation can be seen as a decrease in visibility (Figure 3). The main aerosol optical properties used in **Papers II–IV** are described in Section 2.2.1.

Air pollution can also affect surface properties. In particular, **BC deposition on snow** decreases the surface reflectivity – albedo – and increases absorption by the surface. This warms the surface and may melt the snow, creating a feedback loop, further decreasing the surface albedo (Hansen and Nazarenko, 2004; Bond et al., 2013). This is important especially in the Arctic areas where, for example, BC from forest fires can be transported to snow-covered areas, as was the case in spring 2006, as described in **Paper III** (see also Stohl et al., 2007; Treffeisen et al., 2007).

The largest uncertainties in our understanding of the forcers of anthropogenic climate change lie within complex interactions with aerosols and clouds (Boucher et al., 2013). As aerosols interact with solar radiation, they can affect the thermodynamic conditions in the atmosphere and therefore cloud formation. For example, large amounts



Figure 3: Haze in Beijing, China, on 24 November 2014.

of aerosols in the atmosphere may decrease the amount of solar radiation reaching the surface and thus hinder initiation of convection or formation of low-level clouds. A layer of absorbing aerosols warms the layer and may either hinder or enhance cloud production around the layer. Absorbing aerosols in a cloud may enhance evaporation of cloud droplets.

Aerosols also act as **cloud condensation nuclei (CCN)** and therefore their number and type are important factors affecting cloud formation and properties. Smaller and more numerous cloud droplets are observed in the presence of more CCN. This may lead to brighter and longer-lasting clouds – both leading to a negative radiative effect (Twomey, 1977). An increase in CCN has also been observed to increase cloud cover (Kaufman and Koren, 2006). Much less is known about aerosol particles’ ability to act as **ice nuclei (IN)** and aerosol effects on cold clouds (Fan et al., 2016). In mixed-phase clouds, the smaller droplets due to more CCN may lead to more freezing and consequently more latent heating in the cloud and higher cloud tops. A special case studied in **Paper IV** is aerosol effects on UTH via changes in the microphysics of deep convective clouds. The theory behind this is described in Section 2.2.2.

2.2.1 Basic concepts of aerosol optical properties

The direct effects of aerosol particles on climate depend on their optical properties, i.e. their interaction with solar radiation.

Propagation of radiation in a medium can be expressed by the **Beer–Lambert law** (Seinfeld and Pandis, 1998):

$$\frac{I}{I_0} = \exp(-\sigma_E z), \quad (1)$$

where I_0 is the incoming radiation flux and I is the flux after distance z .

$$\delta = \sigma_E z \quad (2)$$

is the optical depth of the medium. σ_E is the extinction coefficient of the medium. It can be interpreted as atmospheric visibility. σ_E consists of extinction by gases and aerosol particles. If we assume gas interaction with solar radiation to be minor, σ_E can be expressed as **the extinction coefficient** of aerosol particles, σ_{EP} . σ_{EP} consists of scattering and absorption:

$$\sigma_{EP} = \sigma_{SP} + \sigma_{AP}, \quad (3)$$

where σ_{SP} is the **scattering coefficient** and σ_{AP} is the **absorption coefficient**.

Aerosol optical depth (AOD) is a commonly used optical parameter that can, for example, be retrieved by satellite instruments. It is defined as

$$AOD(\lambda) = \int_0^{TOA} \sigma_{EP,\lambda}(z) dz, \quad (4)$$

where z is height from the ground and TOA means top of the atmosphere.

Ångström exponent α describes the wavelength dependency of the aerosol optical properties. For AOD it is defined as

$$\alpha = -\frac{\log \frac{AOD_{\lambda_1}}{AOD_{\lambda_2}}}{\log \frac{\lambda_1}{\lambda_2}} \quad (5)$$

and respectively for scattering and absorption coefficients in **Paper II**. It can be used to qualitatively estimate the size distribution of the particles: the smaller the particles, the larger the α (Ångström, 1929).

Aerosol forcing efficiency $\Delta F/\delta$ can be used to estimate aerosol forcing by unit optical depth. The formula is given in **Paper II**, Eq. 7 or in Sheridan and Ogren (1999).

2.2.2 Aerosols affect upper tropospheric humidity

Aerosol effects on climate via changes in deep convective clouds are studied in **Paper IV**. Aerosol effects on shallow clouds have been studied extensively, yet they are still far from being understood (Boucher et al., 2013). Aerosol effects on deep convective clouds, however, have received attention only very recently (Tao et al., 2012; Rosenfeld et al., 2014; Fan et al., 2016).

According to modelling studies, more CCN available in the updrafts of deep convection may lead to more and smaller droplets, and consequently delayed onset of precipitation (Khain et al., 2005; Fan et al., 2012a). This may lead to a decreased amount of warm rain and increased amount of ice transported to the upper troposphere (Ramanathan et al., 2001; Fan et al., 2010b,a). It was proposed by Ramanathan et al. (2001) that suppression of precipitation in deep convection could thus lead to increased amount of water vapour in the upper troposphere. Bister and Kulmala (2011) argue that aerosols can increase UTH even without changes in precipitation and can thus have a global warming effect on climate. The mechanism is presented in Figure 4a.

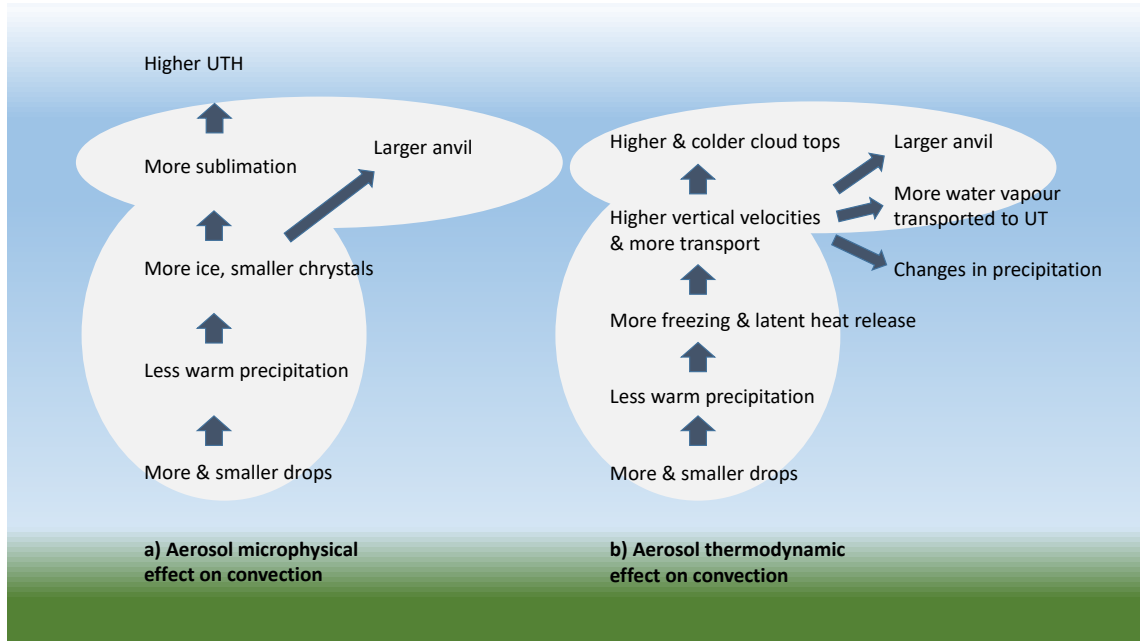


Figure 4: Aerosol effects on deep convective clouds. Aerosol **(a)** microphysical effect and **(b)** thermodynamic effect (also called as invigoration effect) presented separately.

In support of the hypothesis, more extensive ice anvils have been observed in association

with deep convection in polluted cases when compared to clean cases (Koren et al., 2005, 2010b,a). Sherwood (2002b) reports smaller ice crystals in continental deep convective clouds as well as in clouds downwind the continents, where aerosol amounts are larger, as compared to clean maritime clouds. Also, Zhang et al. (2007) report a decadal increase in high cloud amounts in the Pacific storm track associated with increased air pollution. UTH is known to increase as a function of upper level cloud amount (Udelhofen and Hartmann, 1995; Soden, 2004; Chung et al., 2004), and, for example, modelling studies by Wright et al. (2009) demonstrate the importance of the microphysics of deep convective clouds on UTH. High UTH has also been observed to collocate with high AOD (Kottayil and Satheesan, 2015).

Also, Sherwood (2002a) observed smaller ice particles in deep convective clouds associated with biomass burning and concluded this could be the reason behind observed moistening of the tropical tropopause layer and the stratosphere. Moistening of the stratosphere is one of the hot potatoes in climate research currently (Solomon et al., 2010). However, possible relations between stratospheric moisture and UTH will not be discussed here.

More freezing may also lead to more vigorous convection as more latent heat is released for convection due to more condensation. In the literature this has been called as **the invigoration effect of aerosols on convection** (also *the aerosol thermodynamic effect on convection*, in contrast to *the aerosol microphysical effect*) and the mechanism is presented in Figure 4b. The invigoration hypothesis has received strong support from several modelling studies (Khain et al., 2005; Fan et al., 2012a,b; Storer and Van den Heever, 2013). Also, observations show higher clouds (Koren et al., 2008a; Li et al., 2011) and an increased rain center of gravity (Heiblum et al., 2012) associated with more CCN.

Although the invigoration hypothesis predicts more latent heating and precipitation formation in deep convective clouds due to more CCN, it is not obvious if aerosols actually increase or decrease precipitation associated with deep convection (Tao et al., 2007; Khain et al., 2008; Khain, 2009; Ekman et al., 2011). It seems to be strongly dependent, for example, on the vertical wind shear (Fan et al., 2009), atmospheric moisture content (Khain et al., 2008) or the amount of IN (Ekman et al., 2007). Interestingly, observations based on satellite data show consistently higher precipitation rates associated with higher AOD (Koren et al., 2012). These studies, however, have faced criticism due to possible measurement bias, as discussed in Section 3.2.

Even though aerosols may change precipitation patterns, it is important to note that the total amount of precipitation is constrained by the atmospheric and surface energy balances (Grabowski and Morrison, 2011). If surface radiation is assumed unchanged, increased aerosol amounts cannot change precipitation amounts globally, but the possible local changes must be compensated elsewhere. Therefore it is not enough to study a single cloud but rather cloud field properties at a larger spatial and temporal scale (Grabowski and Morrison, 2011; Morrison and Grabowski, 2011).

The invigoration of convection may also lead to locally increased UTH. However, this effect can be only local, as changes in precipitation must be compensated elsewhere in time and space, as stated above. In **Paper IV** we argue that even without the invigoration effect, aerosols can affect UTH by increasing the number of ice crystals (Figure 4a). We eliminated the invigoration effect from our studies by binning the data according to precipitation, i.e. comparing cases with similar amounts of precipitation. Studies by Fan et al. (2013) support the hypothesis by showing that aerosol effects on the deep convective cloud microphysics have an important effect on anvil properties even when the invigoration effect is absent.

Upper tropospheric moisture is governed by deep convective and mid-latitude weather system activity that transports water from the lower to upper troposphere (Held and Soden, 2000; Gettelman et al., 2006). UTH is usually expressed in terms of relative humidity, because it can be derived from the satellite instruments relatively easily (Eq. 6), but also because the absolute amounts of water vapour are so strongly dependent on the vertical temperature gradient of the upper troposphere. In the upper troposphere, high UTH values are observed in the vicinity of deep convective clouds and subsequent anvil clouds (Chung et al., 2004; Udelhofen and Hartmann, 1995). The importance of sublimation of cirrus ice for UTH has been questioned based on the small amounts of ice in the anvil clouds (Horvath and Soden, 2008; Soden, 2004). However, contradictory results have also been shown. For example, Wright et al. (2009) show that eliminating condensate evaporation in a general circulation model causes significant drying of the tropical upper troposphere.

It is actually a bit surprising how little is known about UTH. Obtaining reliable atmospheric soundings has been problematic in cold upper tropospheric temperatures (Elliott and Gaffen, 1991). Also general circulation models as well as reanalysis data sets struggle in getting, in particular, the tropical UTH right (Eriksson et al., 2010; Chung et al., 2013). It is probably because the transport of the moisture happens in the

turbulent eddies generated by moist convection, the scales of which are not explicitly solved in these models (Held and Soden, 2000).

No significant global trends in UTH have been observed since the start of satellite measurements in the 1980s, although absolute humidities have increased with increasing temperatures (Hartmann et al., 2013; Shi and Bates, 2011; Chung et al., 2014). However, Bister and Kulmala (2011) reason that the largest changes in atmospheric aerosol loadings happened well before the satellite era, and also that aerosols could be behind some of the observed local trends in UTH.

The Earth’s radiative transfer is highly sensitive to upper tropospheric water vapour (Held and Soden, 2000). According to Chung et al. (2014), 80 % of the total water vapour feedback of global warming comes from water vapour in the upper troposphere. As the absolute amount of water vapour in the upper troposphere is small, even small changes in UTH due to aerosols would cause a significant warming effect on climate.

The challenge here is that the microphysics of deep convection is poorly presented in climate models (Boucher et al., 2013; Khain et al., 2015). There are two main approaches to include aerosol effects on clouds: bulk microphysics parameterization and spectral (bin) microphysics. Studies show that only spectral (bin) microphysics can capture the aerosol effects on deep convective clouds. However, that is computationally too expensive for the general circulation models that use bulk parameterization schemes. Therefore, aerosol effects on deep convective clouds and UTH are not currently included in the climate change projections produced by the general circulation models.

2.2.3 About meteorological covariation in aerosol–cloud interaction studies

As stated in Section 2.1, atmospheric aerosol concentration is a strong function of meteorology, i.e. prevailing atmospheric conditions. So are clouds. This makes studying aerosol–cloud interactions a complex field. In the following, I describe some of the challenges related to that:

1. Aerosol emissions are often linked to meteorology.

For example, production of sea salt aerosol is governed by winds, and sea salt emissions are known to increase as a function of wind speed (O’Dowd and Smith, 1993). This has been observed to cause positive correlations with AOD and cloud fraction (Engström and Ekman, 2010).

Aerosol emissions also have strong seasonality (Remer et al., 2008). In particular, dust and biomass burning emissions show strong seasonality according to dry and wet seasons. This may cause strong correlations between aerosol and cloud properties. For example, dry seasons are often linked to high dust emissions as well as descending large-scale motion and a dry upper troposphere.

2. Aerosol transport is ruled by meteorology.

Aerosols are transported in atmospheric motions. Meteorological patterns affecting both aerosols and cloud properties have been observed to cause spurious correlations (Grandey et al., 2011, 2013). Examples of the importance of weather systems on aerosol transport are described in **Paper III**.

3. Aerosol removal processes are related to meteorology.

Precipitation removes aerosol particles from the air. As precipitation is obviously related to clouds, it is thus expected to cause negative relations between aerosols and cloud-related properties in regions of frequent precipitation. Several studies have found wet scavenging to obscure aerosol–cloud relations (Grandey et al., 2013; Quaas et al., 2010).

4. Aerosol humidification.

Aerosol particles are known to swell in humid conditions, depending on their hygroscopicity (Swietlicki et al., 2008), leading to an increase in AOD (Jeong and Li, 2010). Aerosol–cloud interaction studies are often conducted using AOD data as an indicator of the number of CCN. As clouds typically form in humid conditions, this has been noted to cause positive correlations between AOD and cloud properties (Quaas et al., 2010; Grandey et al., 2013). More of the biases related to AOD measurements are discussed in Section 3.2.

5. Causalities are complex.

Sometimes, deriving causality becomes complex. It is not an easy task to derive whether aerosols are the cause behind observed changes in cloud properties, or whether some other meteorological factors are behind both observed values, or

to what extent aerosols can modify meteorology, which then controls cloud formation (Stevens and Feingold, 2009). For example, Gryspeerdt et al. (2014a) show that large parts of the observed AOD–cloud top height relationship can be explained by AOD–cloud fraction and cloud fraction–cloud top height relationships. Or the study of Mauger and Norris (2007) shows that more than half of the observed AOD–cloud fraction relationship in the North-East Atlantic was explained by similar features in air mass history that effected the lower tropospheric static stability. Attempts to take into account all possible meteorological factors have been made, for example, by Koren et al. (2010a), Gryspeerdt et al. (2014b) and in **Paper IV**.

3 Research methods

3.1 Ground-based measurements of gases and aerosols

In this thesis, atmospheric measurement data from three ground-based measurement stations in Finland are used: SMEAR I in Värriö, Lapland, SMEAR II in Hyytiälä, southern Finland, and SMEAR III in Helsinki. SMEAR stands for Station for Measuring Ecosystem-Atmosphere Relations and stations are equipped with comprehensive instrumentation to monitor the atmospheric composition as well as its interactions with the surrounding ecosystems.

SMEAR I is a remote measurement station in the middle of Värriö nature park in eastern Finland (69°46'N, 29°35'E), close to the border with Russia (Hari et al., 1994; Ruuskanen et al., 2003). In this thesis, atmospheric SO₂, NO_x, O₃, CO and aerosol particle number size distribution data are used. The measurements are made at different heights from a 15 m tall measurement tower located on the top of a hill 390 m above mean sea level.

SMEAR II is a remote measurement station in the middle of a boreal forest in Juupajoki, southern Finland (61°51'N, 24°17'E; Kulmala et al., 2001b; Hari and Kulmala, 2005). The station has comprehensive equipment to monitor the atmospheric constituents as well as the surrounding ecosystem, for example a 127 m high mast, where atmospheric composition is measured at several heights (Figure 5). In this thesis, SO₂, NO_x, NO, O₃, CO, CO₂, H₂O, and aerosol particle number size distribution data are used, as well as BC concentration and aerosol optical properties measured with a nephelometer and an aethalometer.

SMEAR III is an urban background measurement station located at Kumpula science campus in Helsinki (60°10'N, 24°57'E; Järvi et al., 2009). The measurements are made from a 31 m high measurement tower located on a hill, 26 m above mean sea level. In this thesis, measurements of SO₂, NO_x, O₃, CO and aerosol particle number size distribution are used.

To estimate the impact of air pollution on climate or its role in environmental changes, consistent high-quality long-term measurements are needed. Continuous measurements of atmospheric constituents started at the SMEAR II station in 1996 (Hari and Kulmala, 2005). The unique data set from the SMEAR II station thus provides the longest



Figure 5: Measurement tower in Hyytiälä SMEAR II.

continuous time series of, for example, submicron aerosol number size distributions in the world (Petäjä et al., 2016). Measurements at SMEAR I started in 1991 (Ruuskanen et al., 2003) and at SMEAR III in 2004 (Järvi et al., 2009). SMEAR data can be found and freely downloaded at <https://avaa.tdata.fi/web/smart/smeaar>.

3.2 Satellite-based measurements

AOD (Eq. 4) can be retrieved either from the ground or by satellites. AOD is in general used to describe the 'amount' of aerosol particles in the atmosphere, although the following limitations should be kept in mind:

- AOD does not indicate the aerosol size. Measurement with a certain wavelength reveals optical depth at that wavelength, but it may be that a few large particles

dominate the optical depth and that particles much smaller than the wavelength are not seen at all. Studying the wavelength dependency, for example with the help of the Ångström exponent α , reveals more about the size distribution of the aerosols. Therefore also the aerosol index ($AI = AOD \times \alpha$, Nakajima et al., 2001) has been used in addition to AOD (see e.g. Gryspeerd et al., 2014b), also in **Paper IV**. The most used wavelength for AOD retrievals is 550 nm, also in **Papers III and IV**.

- AOD does not indicate the aerosol type. Therefore, it does not, for example, indicate the aerosol particles' ability to act as CCN or IN. As AOD does not provide information about the aerosol particle composition, it does not provide information about the hygroscopic growth either. Aerosol particles are known to swell in humid conditions (Swietlicki et al., 2008) leading to an increase in AOD (Jeong and Li, 2010). This is especially important to note when studying aerosol–cloud interactions, as clouds typically form in humid conditions (Quaas et al., 2010; Grandey et al., 2013).
- AOD cannot be retrieved through clouds. Usually cloudy pixels are removed from the AOD data in quality control processes. This provides challenges to aerosol–cloud studies where the intention is often to study collocated AOD and cloud properties. This may be solved by assuming AOD to be homogenous below the clouds and in the surrounding areas. The approximation may be better over ocean than over land where sources are more varying.
- Clouds may affect AOD retrievals. Enhanced reflection of radiation near cloud edges may lead to enhanced AOD values (Twohy et al., 2009; Várnai and Marshak, 2009; Redemann et al., 2009; Chand et al., 2012; Várnai et al., 2013). In particular, in broken cloud fields, areas between the clouds may be classified as cloud-free even though the data may be biased by very small clouds (Koren et al., 2008b, 2009). Optically thin clouds are also a challenge for AOD retrievals (Kaufman et al., 2005; Huang et al., 2011, 2012; Chew et al., 2011; Sun et al., 2011; Wollner et al., 2014).
- AOD does not indicate at which height in the atmosphere the aerosols are, except when height-resolving instruments, like lidars, are used. In general, the majority of the AOD comes from the boundary layer, i.e. the lowest ca 1 km of the troposphere, as the aerosol sources mainly are at the surface. However, there

are exceptions, for example when high temperatures associated with intensive burning events lift pollutants to high altitudes (**Paper III**).

These challenges have raised discussion about the reliability of the aerosol–cloud interaction studies based on AOD retrievals (Grandey et al., 2013; Quaas et al., 2010).

In **Paper IV** we used AOD as an indicator of the amount of CCN. Keeping the limitations in mind, AOD has in general been considered a moderate proxy of CCN (Andreae, 2009). In **Papers III** and **IV** we used AOD from the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument onboard the Aqua and the Terra satellites (Remer et al., 2005). In **Paper III** we also used vertical profiles of aerosol optical properties obtained from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite (Winker et al., 2009).

In **Paper IV**, UTH was obtained from the microwave humidity sounder (Bonsignori, 2007) onboard the MetOp-A satellite. The microwave humidity sounder is a scanning radiometer that uses microwave channels of 89–190 GHz to detect atmospheric water vapour at three different heights. UTH is retrieved from the radiances as

$$\ln(UTH) = a + bT_b \quad (6)$$

where T_b is the radiance expressed as brightness temperature and a and b are constants given in Buehler and John (2005). UTH represents the mean relative humidity with respect to water between 500 hPa and 200 hPa. The accuracy of the method is estimated to be 2 %RH and 7 %RH for low and high UTH values, respectively. The microwave method developed by Buehler et al. (2008) enables to study also in the areas of the anvil clouds.

Precipitation data from the Tropical Rainfall Measuring Mission (TRMM, Huffman et al., 2007) was used in **Paper IV** to compare cases with similar amounts of precipitation in order to eliminate the effect of invigoration of convection due to aerosols. The TRMM daily rainfall product was used, consisting of multiple satellite as well as in situ measurements during a period of 24 h.

3.3 Trajectory analysis

Analysis of atmospheric transport is needed when the intention is to understand where the measured concentrations come from. This is important, for example, in the case of exceptionally high concentrations, as in **Paper III**, when information on the potential sources is critical in order to prevent harmful health effects. Also, understanding air pollution climatology is essential in air quality modelling.

Atmospheric transport of measured compounds can be approximated as a first guess by using measured wind data (direction and speed) or other meteorological data, like atmospheric soundings. For a more comprehensive look there are several atmospheric reanalysis data sets available where measurements around the globe are assimilated into atmospheric models. Based on these data, the atmospheric transport can be calculated. The simplest way to estimate the transport is to calculate backwards in time the most probable path for an air parcel arriving at the measurement station at a certain time. By doing so we get a **backward trajectory** for the air parcel. A single trajectory does not take into account any mixing with the surrounding air or the uncertainty in the transport modelling so it is always a crude approximation. For more accurate estimations, advanced modelling, like FLEXPART (FLEXible PARTicle dispersion model; Stohl et al., 2005) or SILAM (System for Integrated modeLLing of Atmospheric coMposition; Sofiev et al., 2006), should be used. However, trajectories are simple and require far less computational capacity. If trajectories are used in large amounts, as described in **Paper I**, these statistical uncertainties can be assumed to smooth out (Stein et al., 2015).

In **Papers I–III** we used the HYSPLIT 4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 1998; Stein et al., 2015) to calculate backward trajectories for the measurement sites. Trajectories were calculated for an arrival height of 100 m above ground level at hourly intervals, going 96 h back in time. At each time step the measured concentration value at the station (Figure 6 i) was assigned to the grid cells ($1^\circ \times 1^\circ$) along the corresponding back trajectory (Figure 6 ii–iv). Thus a concentration field is acquired, where values in each grid cell describe the mean concentration measured in the air arriving at the measurement station from that location (Figure 6 v). In **Papers I–III** also the horizontal uncertainty of the trajectories was taken into account by weighting the concentrations with the trajectory uncertainty.

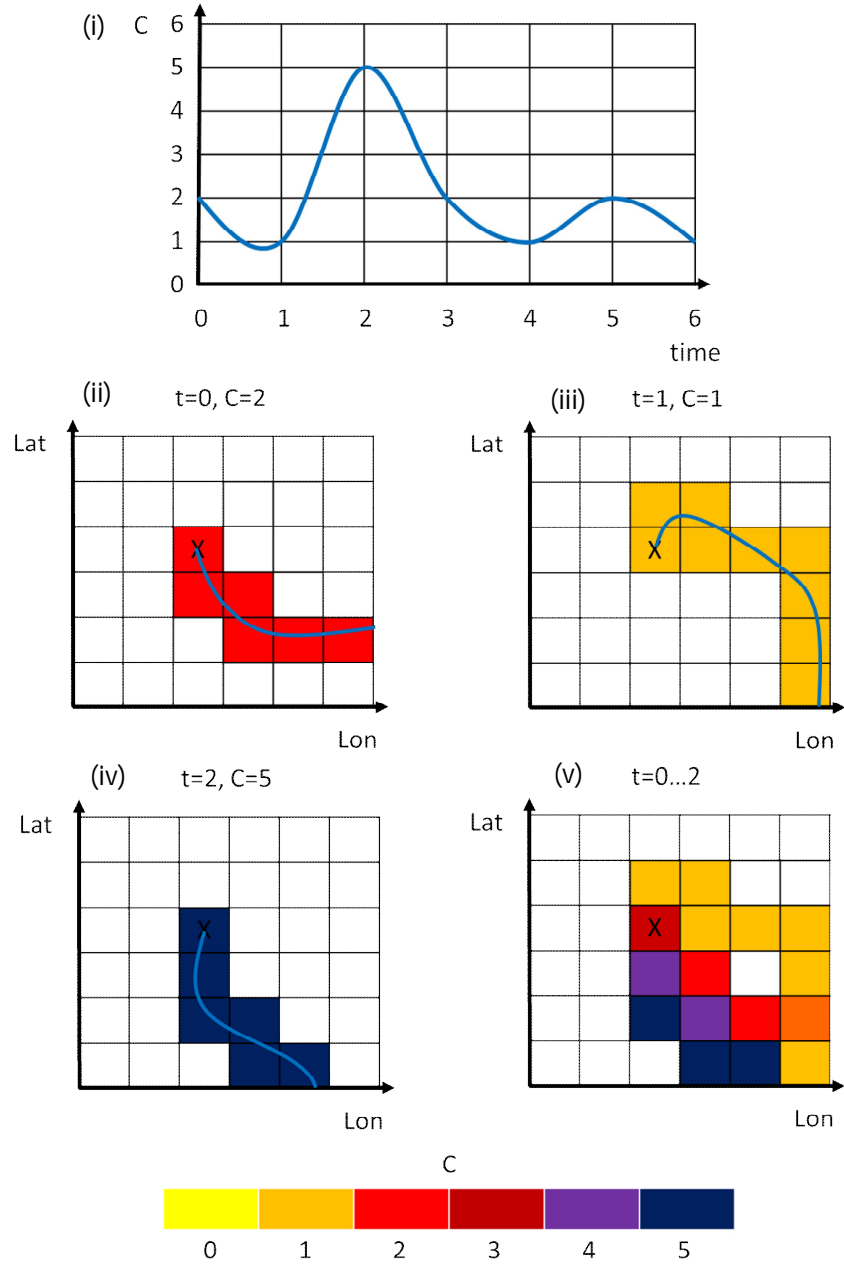


Figure 6: **(i)** Concentration (C) as a function of time measured at the measurement station. **(ii–iv)** Measured concentrations (C) at each time step (t) in **(i)** are attached to air mass trajectories calculated for the same time step. Trajectories are in a latitude-longitude grid. The concentration field in **(v)** is obtained as an average of the fields in **(ii–iv)**.

The method described above was originally introduced by Ashbaugh et al. (1985) and named later as Potential Source Contribution Function (PSCF). Variations of the method have been used widely to study air pollution transport (Fleming et al., 2012), for example in Southern Africa (Vakkari et al., 2011), the Alps (Kaiser et al., 2007) or Lapland (Aalto et al., 2002). Exactly the same method as in **Papers I–III** has been used by Chen et al. (2016) to study the transport of radon to Finland.

4 Results

4.1 Transport and properties of air pollutants in Finland

Hyttiälä is in general seen as a relatively clean background station. The median concentrations of anthropogenic pollutants are very low: 1.3 ppb for NO_x and 0.2 ppb for SO_2 (**Paper I**, Table 1). Therefore, the anthropogenic contributions can be traced from the data. The 99-percentiles of NO_x and SO_2 are 10.0 ppb and 4.4 ppb, respectively.

Trajectory analysis (**Paper I**) revealed the southern and eastern origin of high air pollution concentrations measured in Finland. High concentrations of accumulation mode particles, SO_2 , and NO_x were observed in south-eastern air masses of continental origin. These transport directions dominated the concentration fields during all seasons. Concentrations of SO_2 and NO_x in air masses coming from the south-east were especially high in winter, whereas concentrations of accumulation mode particles were highest in south-eastern air masses in summer. Western and northern directions were relatively clean in terms of these pollutants, despite a contribution of SO_2 and NO_x from the north also in winter. Although there are point sources of these constituents in Finland, as named in **Paper I**, this strongly suggests that emissions from industrial sources in Eastern Europe and Russia accumulate in these continental air masses that often get transported to Finland when there is a high pressure system east of Finland, as was the case in April–May 2006, described in **Paper III**.

Interestingly, the evident pollution sources in western Europe were clearly under-represented in the Hyttiälä measurements. This is probably due to the different climatology of these air masses, as discussed in **Paper I**: there is more rain and wet removal when the air comes from the west, for example in connection with low pressure systems of Atlantic origin.

The smallest aerosol particles (nucleation mode, 3–25 nm) showed different phenomenology: the highest concentrations were measured in air masses of north-western, i.e. clean Atlantic, origin. The same feature has been observed in several studies (e.g. Sogacheva et al., 2005; Tunved et al., 2005, 2006) that new aerosol particles are formed from biogenic precursor gases in clean air masses where the preceding aerosol load (the CS) is small. When the air is polluted, the precursor gases condensate (or the newly formed particles coagulate) on the surfaces of the existing aerosols rather than form new

ones. Therefore the highest concentrations are observed in north-western air masses especially in spring (March–May) and in September, when the new particle formation mechanisms are strongest. Therefore the total fine mode particle number concentration (and also Aitken mode particle concentration) shows a bidirectional pattern (Figure 7).

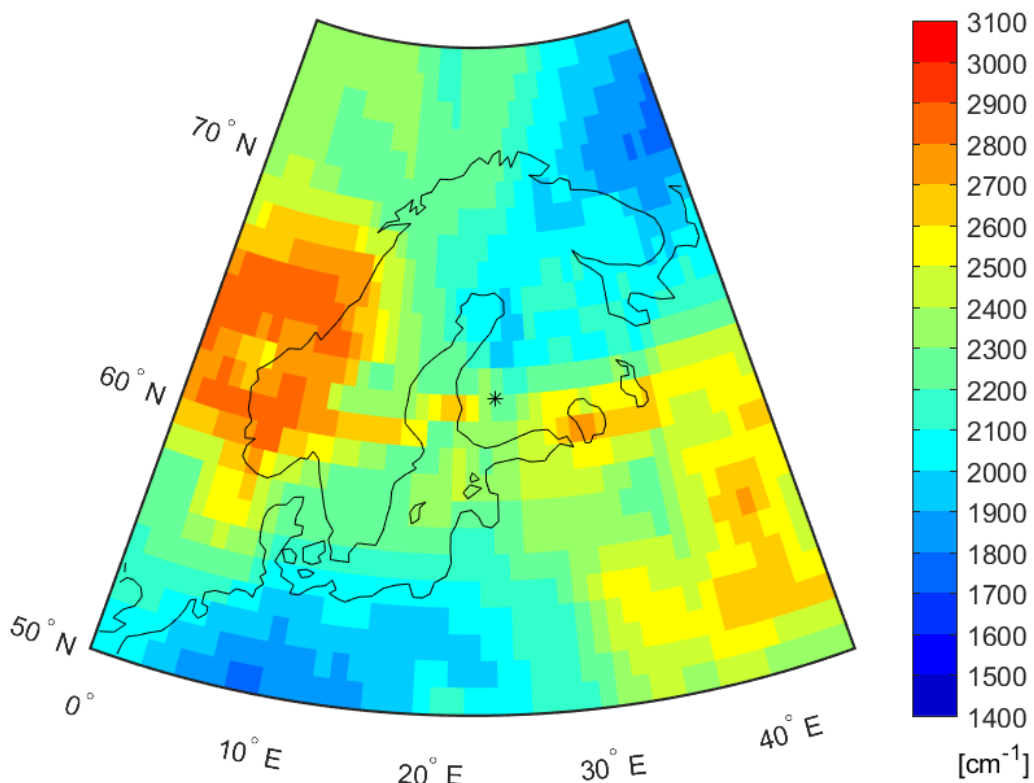


Figure 7: The concentration field for average total fine particle number concentration (3–1000 nm) in Hyytiälä in 1996–2008.

Trajectory analysis also allowed us to study decadal changes in concentrations coming from different sectors. A general decrease in SO_2 and NO_x concentrations is observed in all sectors between years 1996 and 2008. Ozone levels, however, show a slight increase in all sectors. Also, particle number concentrations in the 25–1000 nm size range has increased in the continental sector between 1996 and 2008. More details about the local trends in Hyytiälä have been reported by Nieminen et al. (2014).

In conclusion, the trajectory statistical method represented in **Paper I** was found to be a useful tool in estimating the contribution of different sectors and transport patterns to measured concentrations at the receptor site. It gave valuable information

on the relative importance of different source areas to the measured concentrations. On the other hand, it was not able to reproduce actual emission sources, and the concentration fields produced based on the back trajectories should therefore not be considered as emission fields. Even if the wet and dry removal of pollutants, chemical transformation and photochemistry as well as dispersion had been taken into account, it is still not likely that the back trajectory method could produce the emission fields, especially as the sources close by would anyway conceal anything that stays behind them. For more accurate purposes, advanced modelling, like FLEXPART (Stohl et al., 1998, 2005), should be used. However, if no supercomputers are available, or as a first approximation, this simple trajectory method is found to be a useful tool.

4.1.1 Optical properties

Aerosol optical properties were studied in **Paper II** at the SMEAR II station, Hyytiälä, Finland, from October 2006 to May 2009.

The scattering coefficient σ_{SP} ($\lambda = 550$ nm) in Hyytiälä on average was 18 ± 20 Mm⁻¹ (average \pm standard deviation). There were not very strong seasonal or diurnal cycles, but the wind direction and trajectory analysis revealed a strong dependency on transport directions: the highest values were coming to Finland from the south-east (24 ± 21 Mm⁻¹ when the wind was blowing from 120°) and the lowest values from the north-west (8.8 ± 9.4 Mm⁻¹ when the wind was blowing from 300°).

The absorption coefficient σ_{AP} ($\lambda = 550$ nm) in Hyytiälä was 2.2 ± 2.4 Mm⁻¹. The seasonal cycle of absorption was stronger than that of scattering, being the highest in winter (2.7 ± 2.5 Mm⁻¹) and the lowest in summer (1.4 ± 1.2 Mm⁻¹). The diurnal cycle of σ_{AP} was stronger than that of σ_{SP} , being the largest in summer when the lowest values of σ_{AP} were observed in the afternoon and highest values during the night.

The wind direction and trajectory analysis showed a similar pattern of the sources of absorbing than scattering aerosols. South-eastern transport routes favoured high absorption coefficients (3.1 ± 2.7 Mm⁻¹ when the wind was blowing from 120°), whereas north-western transport routes brought low absorption coefficients (1.0 ± 1.2 Mm⁻¹ when the wind was blowing from 300°).

The Ångström exponent of scattering, α_{SP} , was 1.7 ± 0.5 . The highest values were ob-

served in spring and summer, indicating the dominance of small particles. The smallest α_{SP} values were measured in winter, suggesting the dominance of large particles. The diurnal cycle was not very strong. There was not such a strong wind or transport direction dependency for α_{SP} as there was for σ_{SP} and σ_{AP} . The lowest α_{SP} , however, came to Hyytiälä from the west. This may suggest dominance of sea salt in western air masses, whereas more anthropogenic influence and variety of aerosol size distributions is to be observed in the eastern air masses.

4.1.2 Biomass burning episodes

The biomass burning episodes in 2006 and 2010 in Hyytiälä and Helsinki, and also the 2006 spring episode in Värriö, clearly stand out from the data as being among the highest air pollution cases in the history of summer measurements at these stations (**Paper III**). The most obvious signs of a biomass burning episode were elevated CO and BC concentrations, as well as CS, total particle volume concentration and number concentration of particles in the accumulation mode size range. Also CO₂, SO₂, NO_x and O₃ had elevated values during some of the episodes, but their correlations with the obvious biomass burning emission tracers CO and BC varied. Because of the positive correlation with BC, we concluded that the elevated concentrations of CO, CO₂ and O₃ were dominantly caused by biomass burning. However, there are also other sources of these pollutants, especially SO₂ and NO_x, in the same areas from where the smoke was transported, which may cause elevated concentrations compared to the reference period.

The particle number size distributions peaked in accumulation mode size range at all three stations during the smoke episodes. The median particle size was 44–84 nm larger during the smoke days than during the reference period. Also the number of particles in the nucleation and Aitken mode size range was reduced. The smoke during the episodes in 2010 was observed to be strongly scattering, but also the absorption coefficients were high, as compared to **Paper II**. Also the Ångström exponents of scattering as well as absorption were high compared to those represented in **Paper II**. This indicates strong secondary particle production in the smoke plume as well as condensation of organic matter onto the particles. The smoke particles were also observed to grow in size during their transport through Finland.

The difference between the smoky air and surrounding air masses was clearly seen also

in the relative source contribution fields calculated for summer 2010 in the same way as in **Paper I**. These fields confirmed the source of these extremely high concentrations to be in the same areas as the biomass burning at that time.

Satellite detections of the AOD revealed the total extent of the smoke as well as the heterogeneous distribution of it. The same was observed with the flight measurements: although elevated particle number concentrations were observed on 29 July 2010 throughout the lower troposphere, the densest plumes were observed around Jämsä city at 2500–3000 m altitude.

We concluded that the composition of the smoke varied between and within the plume days, indicating differences in the sources, burning conditions and chemical transformations taking place during the atmospheric transportation of the plumes.

4.2 Direct effects of air pollutants on climate by scattering and absorption of solar radiation

Based on **Paper II**, aerosols observed in Hyytiälä were more scattering than absorbing, meaning that they more cooled the climate system than warmed it. The average forcing efficiency was $-23.0 \pm 5.0 \text{ W m}^{-2}$. Aerosols observed in summer cooled the climate system more than aerosols observed in winter. However, if the surface was covered with snow, the aerosols observed in winter eventually heated the climate system.

The contribution of submicron particles to scattering in Hyytiälä was $\sim 90 \%$. It was largest in winter (92 %) and smallest in summer and autumn (88 %). The average contribution of particles smaller than 100 nm to total scattering was 0.2 %.

Also the biomass burning smoke observed in 2006 and 2010 was highly scattering, although the smoke also contained large amounts of absorbing material (BC). We can conclude that the smoke in the air cooled the atmosphere. However, in snow-covered areas the case may be the opposite, as stated in Section 4.1.1. Also, BC deposition on snow may decrease surface albedo, having a warming effect on climate (Hansen and Nazarenko, 2004). In 2006 the smoke was transported high into the Arctic, to the snow-covered areas in Lapland and Spitsbergen (Treffeisen et al., 2007; Stohl et al., 2007).

4.3 Indirect effects of air pollutants via changes in cloud microphysics: aerosols increase upper tropospheric humidity

In addition to direct effects on the Earth’s radiation budget, aerosols have many complex and often nonlinear effects on climate via changes in cloud properties (Fan et al., 2016). In **Paper IV** we show first observational evidence of a so-far overlooked effect of aerosols on deep convective clouds that increases UTH. The theory behind this is described in Section 2.2.2.

By comparing cases of $\text{AOD} \geq 0.18$ and $\text{AOD} < 0.18$ over the ocean East of China, we observed UTH to be on average 2.2 ± 1.5 %RH larger with large AOD. We used only values when precipitation was more than 1 mm day^{-1} . Different data products, spatial averaging, time span of the measurements, as well as AOD limits were tested. In general, a higher UTH was observed with a higher value of AOD, although the magnitude of the change depended on specific choices.

We made an effort to exclude all other possible causes of the UTH increase than the aerosol effect on deep convective cloud microphysics. Meteorological covariation and aerosol humidification have been noted to obscure causalities in aerosol–cloud interaction studies (Section 2.2.3). Binning the data according to precipitation should already alleviate these problems, but in addition to that we also binned the data according to seven meteorological parameters of ERA Interim reanalysis data (Dee et al., 2011). These parameters were: mean sea level pressure, pressure change from previous day, 550 hPa omega, low cloud cover, 2-m relative humidity, 10-m wind speed and 10-m wind direction. If meteorological covariation or aerosol humidification would explain the UTH increase associated with large AOD, binning by lower tropospheric humidity or other meteorological variables should significantly decrease the observed UTH change. However, practically the same values were observed as when binning only by precipitation. Only with extreme values of some meteorological parameters, which are likely associated with the recent passage of low pressure systems with wet scavenging dominating the AOD as discussed in **Paper IV**, no change or even a decrease in UTH was observed. Therefore we concluded that meteorological covariation or aerosol humidification are not the causes of the observed UTH increase.

It has also been noted that MODIS AOD retrievals may get contaminated by simultaneous cirrus (Kaufman et al., 2005; Huang et al., 2011, 2012; Sun et al., 2011; Wollner et al., 2014). In our study region we observed AOD to increase almost linearly as a

function of the cirrus fraction. To make sure that our results are not biased by cirrus contamination, we removed this dependence from the AOD data by linear regression. However, by doing this, we may have removed part of the real microphysical effect of aerosols on UTH. Namely, more extensive cirrus anvils have been observed in association with higher AOD (Koren et al., 2005; Fan et al., 2013). If we did not remove the AOD dependence on cirrus, the UTH change for $\text{AOD} \geq 0.18$ and $\text{AOD} < 0.18$ was 5.8 ± 1.4 %RH – much larger than if we removed the effect.

Radiative transfer calculations for tropical reference soundings show that even relatively small changes in UTH have a large effect on the outgoing long-wave radiation. For example, 2 %RH increase in UTH in tropical moist air causes a positive radiative effect of 0.50 W m^{-2} . In mid-latitude dry air the effect is even larger, 0.77 W m^{-2} . For 5 %RH increase in UTH the radiative effect was 1.22 W m^{-2} for tropical moist and 1.86 W m^{-2} for mid-latitude dry air. These values are high if we consider the general estimates of the radiative effect of anthropogenic aerosols. Boucher et al. (2013) have estimated the globally averaged effective radiative forcing due to anthropogenic aerosols to have a best estimate of -0.9 W m^{-2} with a wide uncertainty range from -1.9 W m^{-2} to -0.1 W m^{-2} . This estimate combines a direct forcing of -0.35 [-0.85 to $+0.15$] W m^{-2} with a forcing due to aerosol-cloud interactions of -0.45 [-1.2 to 0.0] W m^{-2} . This raises the question of whether anthropogenic aerosols have had a warming effect on global climate since the industrial revolution, in addition to their direct cooling effect, as stated by Bister and Kulmala (2011). If so, the climate sensitivity to increased amounts of greenhouse gases would be smaller than estimated.

5 Review of papers and the author's contribution

Paper I investigates atmospheric transport of fine particles, SO_2 , NO_x and O_3 to a measurement station in Finland with a statistical method based on air mass trajectories and 13 years of continuous ground-based measurements. I wrote the trajectory statistics code, performed the final runs and wrote the final version of the paper.

Paper II reports aerosol optical properties at a boreal research station. I was responsible for the trajectory statistical analysis, together with Mira Hulkkonen. We conducted the trajectory study and wrote the corresponding part of the paper.

Paper III studies some of the most intense biomass-burning smoke episodes in Finland with surface, airplane and remote sensing measurements. I was responsible for the satellite data analysis, meteorological analysis as well as transport analysis of the study. The study was designed and the results discussed together with other authors. Katri Leino wrote the initial version of the manuscript. I was responsible for the writing of the published version as well as the final figures.

Paper IV investigates a new proposed mechanism of aerosol–cloud interactions via changes in the microphysics of deep convective clouds. With satellite data the study shows observational evidence for aerosols increasing UTH. I designed the study and wrote the paper mainly together with Marja Bister. I conducted all of the studies, except for the radiative transfer runs, which I designed but were conducted by Anu-Maija Sundström.

6 Conclusions

Climate change and air quality are the big challenges of our time. They will remain such for some time, because they are highly complex issues. The big challenges cannot be tackled without multidisciplinary understanding of the smallest of the small, like formation of new aerosol particles or how they affect cloud microphysics, as well as the big picture: the general laws ruling our planet's climate system.

These studies highlight the importance of using multiple methods in the analysis of the climate system and the factors influencing it. In my thesis work I have used satellite data of aerosol optical properties, precipitation, clouds and UTH, in situ measurement data of gases and aerosols at three measurement stations, combined with flight measurements, air mass trajectories, synoptic maps and meteorological reanalysis data.

In this work the optical properties, concentrations, as well as favourable transport routes of air pollutants in southern Finland were characterized. It was found that the south-eastern transport routes with continental origin on average bring the highest concentrations of anthropogenic gaseous and particulate pollution to southern Finland. The air masses coming from the north-east were found to bring air that is clean of anthropogenic pollution but with high concentrations of nucleation and Aitken mode particles. Air mass trajectory statistical methods were found to be useful tools to study air pollution transport, but they were not able to reproduce emission source maps.

The most severe air pollution cases in the history of air pollution measurements in Finland were produced by the biomass burning events in Eastern Europe in 2006 and 2010. The smoke was transported hundreds and even thousands of kilometres and the smoke particles were observed to grow in size during transportation.

Particulate air pollution in southern Finland was found to scatter solar radiation and have a cooling effect on climate. High absorption coefficients were found in continental air pollution from Eastern Europe as well as in biomass burning smoke. However, also these air pollutants were on average more scattering than absorbing.

The most important finding in this thesis was related to the aerosol effect on UTH. Based on satellite observations we were able to show the first observational evidence with strong emphasis on causality that aerosols increase UTH and have thus a so far overlooked warming effect on climate. The magnitude of the observed increase is so big that the mechanism potentially has relevance for the global climate.

Aerosol effects on deep convective clouds have potentially large undiscovered and unquantified effects on climate. In particular, the aerosol effect on UTH is definitely worthy of future study. It is also to be noted that the mechanism presented in Section 2.2.2 may not be limited to deep convective clouds but implications for UTH via large-scale precipitation may exist, too. Also, air pollution climatology should be applied more than currently is done in aerosol–cloud interaction studies in order to lessen the meteorological covariation bias in studies.

Studying aerosol–cloud–climate interactions is challenging. This is because aerosols, clouds and radiation are strongly interlinked and the interactions are often far from linear. Aerosol is a strong function of meteorology: air mass history and origin as well as cloud and precipitation processes determine atmospheric aerosol loadings after certain emissions. These factors still are the main drivers of our climate and the hydrological cycle – the major drivers of human life conditions. Studying them and understanding them will remain a fundamental challenge of climate science for the next decades.

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